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INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference HMJ03681WO		FOR FURTHER ACTION		See Form PCT/PEA/416
International application No. PCT/EP2004/004687		International filing date (day/month/year) 26.04.2004	Priority date (day/month/year) 24.04.2003	
International Patent Classification (IPC) or national classification and IPC B29C55/06, B29C67/24, B29C47/36				
Applicant RASMUSSEN, Ole-Bendt				
<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 7 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input checked="" type="checkbox"/> sent to the applicant and to the International Bureau a total of 32 sheets, as follows:</p> <p><input checked="" type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).</p> <p><input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.</p> <p>b. <input type="checkbox"/> (sent to the International Bureau only) a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>				
<p>4. This report contains indications relating to the following items:</p> <p><input checked="" type="checkbox"/> Box No. I Basis of the opinion</p> <p><input type="checkbox"/> Box No. II Priority</p> <p><input type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p><input type="checkbox"/> Box No. IV Lack of unity of invention</p> <p><input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p><input type="checkbox"/> Box No. VI Certain documents cited</p> <p><input type="checkbox"/> Box No. VII Certain defects in the international application</p> <p><input checked="" type="checkbox"/> Box No. VIII Certain observations on the international application</p>				
Date of submission of the demand 11.11.2004		Date of completion of this report 30.06.2005		
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465		Authorized Officer Attalla, G Telephone No. +49 89 2399-6004 		

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/EP2004/004687

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
 - ☐ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
 - ☐ international search (under Rules 12.3 and 23.1(b))
 - ☐ publication of the international application (under Rule 12.4)
 - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report):*

Description, Pages

2, 4, 7, 10, 11, 13, 15, 16, 18-20, 23-25, 27, 29-31, 34-39, 41-43 as originally filed

1, 3, 5, 5a, 6, 8, 9, 12, 14, 17, 21, 22, 26, 28, 32, 33, 40 received on 11.11.2004 with letter of 08.11.2004

Claims, Numbers

1-97 received on 11.11.2004 with letter of 08.11.2004

Drawings, Sheets

1/18-18/18 as originally filed

- ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. ☐ The amendments have resulted in the cancellation of:
 - ☐ the description, pages
 - ☐ the claims, Nos.
 - ☐ the drawings, sheets/figs
 - ☐ the sequence listing (*specify*):
 - ☐ any table(s) related to sequence listing (*specify*):
4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
 - ☐ the description, pages
 - ☐ the claims, Nos.
 - ☐ the drawings, sheets/figs
 - ☐ the sequence listing (*specify*):
 - ☐ any table(s) related to sequence listing (*specify*):

* If item 4 applies, some or all of these sheets may be marked "superseded."

**INTERNATIONAL PRELIMINARY REPORT
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Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-97
	No: Claims	
Inventive step (IS)	Yes: Claims	1-97
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-97
	No: Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item V.

1. Claim 1 discloses a method of manufacturing an oriented film from a blend of two polymers having different melting points, comprising stretching the blend partly when both polymers are melted and partly when only the one with lower melting point is melted. The method allows to obtain fibrils of the polymer with higher melting point dispersed in the other polymer, the fibrils being oriented in one direction and having width and thickness lower than 5 μm .

None of the documents of the available prior art discloses or render obvious the method of claim 1. Therefore the subject matter of this claim is considered to fulfil the requirements of Art. 33 PCT.

2. Independent claims 67 and 73 disclose apparatuses specifically conceived for producing a stretched film according to the method of claim 1, which comprise first means for stretching the extruded film at high temperature and second means for stretching the film at lower temperature and means for cooling the film between the two stretching steps, said means comprising a frictional device. For the same reasons as in point 1 above, also the subject matter of these claims is considered to fulfil the requirements of Art. 33 PCT.

3. Document US-A-2262989 (D1) discloses a method for forming a sheet of thermoplastic polymers (cf. page 3, left col., line 9-13) by extrusion through a die, in which the flow passage through the die comprises an exit orifice having an exit gap (slot orifice 55), whereby upstream from the exit orifice there is provided a grid chamber (space 49 and cavity 50) comprising a grid (screen 52) through which the polymers pass, the grid being located at a position in the chamber where the gap is wider than said exit gap (cf. fig. 8), the grid chamber further comprising a gap reduction portion between the screen and the die exit (cavity 50; cf. fig. 8) wherein the gap through which the polymers flow is reduced at least part way to the gap of the die exit (slot orifice 55). D1 discloses that the method can operate on "semiplasticized or mechanical mixtures of fine particles of cellulose ethers, esters or resins and their respective plasticizers" (cf. page 3, left col., line 11-13) and that the "material is further commingled and compressed in a chamber 49 from whence it is forced through screens 52 which further mixes and filter the material" (cf. page 4, left col.,

line 25-28). Therefore a person skilled in the art finds in D1 the teaching that the method is suitable to obtain an intimate blend of polymers in which the components are well commingled. The subject matter of claim 32 differs from D1 in that it specifies that the grid consists of at least four lamellae, between which, apertures of a size selected to reduce the average size of the polymer dispersion are provided. Neither in D1 nor in any other piece of the available prior art a grid as defined in claim 32 is disclosed or even only suggested. Also the subject matter of this claim is considered to meet the requirements of Art. 33 PCT.

4. Document EP-A-337719 (D3) discloses an extruded oriented film comprising a layer of alloy of two polymers (second resin (corresponding to P1) and first resin (corresponding to P2)) both partly crystalline under 100 °C (e.g. nylon 6 (P1) and polyethylene (P2), as in example 2), wherein P2 in its unoriented state at 20 °C exhibits a coefficient (modulus) of elasticity more than 15% lower than P1, and the alloy comprises a dispersion of microscopically fine fibrils of P1 surrounded by P2, wherein each fibril extends mainly in one direction (cf. page 2, line 49-52; fig. 1) and has width and thickness lower than 5 µm (cf. page 3, line 5-6), whereby the fibrils are flat and generally parallel with the main surfaces of the film (cf. page 2, line 52) with thickness lower than 1 µm (in the range 0.01 µm to 5 µm) and width more than five times the thickness (0.1 µm).

The expression "the oriented film exhibits location of rupture of the P1 fibrils, which locations extend in a generally linear fashion at an angle to the direction of orientation" (cf. also objection in point 2.4) only describes a result to be achieved and cannot be used to distinguish the subject matter of the claim over the prior art. The subject matter of claim 49 differs from D3 in that the film is crosslaminated to another oriented film or is in the form of a rope, twine or woven tape. As D3 does not disclose or give any hint to use the oriented film as claimed in claim 49, the subject matter of this claim is considered to meet the requirements of Art. 33 PCT.

5. D3 additionally discloses that P1 (the second resin) is chosen to exhibit desirable barrier properties (cf. page 2, line 50-51). Then the subject matter of claim 56 differs from D3 for the additional feature that at least four die lines (as defined in the paragraph bridging pages 29 and 30 of the description). Such a feature is not present in the film of D3. Therefore the subject matter of this claim is considered to meet the

requirements of Art. 33 PCT.

6. The subject matter of claim 60 is distinguished from D3 for the additional feature that the film is expanded. As there is no indication in D3 that the film having the fibrillar structure could also be expanded, the subject matter of claim 60 is considered to meet the requirements of Art. 33 PCT.
7. Claim 89 discloses an apparatus specifically designed to carry out the method of claim 32. Therefore, for the same reasons as in point 3 above, also the subject matter of this claim is considered to meet the requirements of Art. 33 PCT.
8. Claims 2 to 31, 33 to 48, 50 to 55, 57 to 59, 61 to 66, 68 to 72, 74 to 88, 90 to 97 are dependent upon claims which meet the requirements of Art. 33 PCT and, as such, also meet the requirements of the PCT with respect to novelty and inventive step.

Re Item VIII.

1. The application contains more than one independent claim per category, namely method claims 1 and 32, product claims 49, 56 and 60 and apparatus claims 67, 73 and 89. The aforementioned claims therefore lack conciseness, as required under Art. 6 PCT. Moreover, lack of clarity as a whole arises, since the plurality of independent claims makes it difficult, if not impossible, to determine the matter for which protection is sought and places an undue burden on others seeking to establish the extent of the protection.
2. Additionally the application does not meet the clarity requirement of Art. 6 PCT for the following reasons:
 - 2.1 In claim 1 it is not clear what is meant by "mechanically determined melting point".
 - 2.2 In claim 49 the meaning of the expression "the oriented film exhibits location of rupture of the P1 fibrils, which locations extend in a generally linear fashion at an angle to the direction of orientation" is not clear.

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(SEPARATE SHEET)**

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2.3 In claim 56 it is not clear what it is meant with "die lines".

11. 11. 2004

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CLAIMS

(62)

1. A method of manufacturing an oriented film by forming a blend of at least two polymers P1 and P2, which both are at least partly crystalline at a temperature below 100°C, whereby P1 has a mechanically determined melting point which is at least 20°C higher than the mechanically determined melting point of P2, extending the blend to form a film and stretching the film, whereby the polymers are selected such that they exist as separate phases in the stretched film, and P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1, and oriented film comprises an alloy of the polymer which is a dispersion of fibrils of P1 surrounded by P2, whereby each fibril extend mainly in one direction and generally has width and thickness which as a mean of these two dimensions is around or lower than 5µm, and further stretching of the film partly takes place by draw-down after extrusion of film while both components are at least partially molten, and partly at a later stage characterised in that the film after said draw-down is hot stretched while P1 is in solid state and P2 is molten or semimolten to selectively orient P1, such that the elongation at break in the direction of this hot stretching, determined by slow drawing at 20°C, is at least 25%, this hot stretching being carried out by drawing the film over a frictionally withholding device.

2. A method according to claim 1, characterised in that after said hot stretching the film is further stretched while both components P1 and P2 are solid, preferably in such a manner that the product film has an elongation at break at 20°C (by slow drawing) of at least 25% in any direction.

3. A method according to claim 1 or claim 2, characterised in that the fibrils are flat with average thickness generally around or lower than 1 µm, preferably generally around or below 0.5 µm, and more preferably generally around or lower than 0.1 µm, and with average width generally around or lower than 5 µm.

4. A method according to claim 1, characterised in that in order to reduce the cross dimensions of the fibrils, the molten blend during extrusion is passed through at least one screen or grid located in a chamber immediately upstream of the exit orifice of the extrusion device, said
5 chamber having a gap higher than the gap of the exit orifice.

5. A method according to claim 4, characterised in that each such grid has walls extending several millimetres in the direction of the flow of the molten blend.

6. A method according to claim 5, characterised in that the major
10 walls in each such grid are slanted so that each forms an angle between about 10 to about 70° to the major surface of the flow entering the grid.

7. A method according to claim 6, characterised in that the slanting and the wall thickness and distances between the walls are such that in a longitudinal section of the device perpendicular to major surface of
15 the flow as this enters the grid, there are at least four such walls.

8. A method according to claim 6, characterised by at least two grids, where in the such walls of one grid are slanted in the opposite direction to the walls of the other grid.

9. A method according to claim 1, characterised in that in
20 succession to the extrusion and attenuation of the blend while both P1 and P2 are molten, the film is first cooled to solidify P1 and P2, thereafter the film is heated in air-lubricated engagement with a heating body of controlled temperature to melt or at least partially melt P2, while keeping P1 solid, and immediate thereafter, while P2 still is at least partially molten and P1 is solid,
25 the film is subjected to the said selective orientation of P1 and subsequent solidification of P2.

10. A method according to claim 1, characterised in that the frictional withholding device comprises one or more bars with rounded edges over which the film is dragged while following an adjustable arc of the bar
30 edge, and said bar or bars are maintained at a temperature which prevents

the film from sticking to the edge or edges, and the length of travel in contact with the edge or edges is adapted to prevent P2 from wholly solidifying.

11. A method according to claim 9, characterised in that at least the process steps from and including extrusion to and including the solidification of P2 are carried out in-line, whereby the line also comprises a hold-back device acting between the cooling and the subsequent heating, and preferably the process steps following solidification of P2 are also carried out in-line with the former process steps.

12. A method according to claim 11, characterised in that the film is extruded as a flat film, and the controlled hold-back between cooling and subsequent heating is established by a roller arrangement, which also may supply the said cooling.

13. A method according to claim 11, characterised in that the film is formed and treated in tubular form from extrusion and at least to the final solidification of P2, whereby the controlled hold-back between cooling and subsequent heating is established by one or more circular rings with rounded edges over which the film is dragged while following an adjustable arc of the rounded edge, and said ring or rings are maintained at a temperature which prevents the film from sticking to the said edge or edges.

14. A method according to claim 11, characterised in that the said heating is carried out with the film in air-lubricated engagement with two heating bodies of which is provided one on each side of the film, the spacing between said heating bodies preferably being adjustable.

15. A method according to claim 1, characterised in that the film immediately after the extrusion is cooled to solidification of P1 while P2 is kept molten or semimolten, and further in immediate succession, the selective orientation of P1 over a frictionally withholding device is carried out with the polymers in such states.

16. A method according to claim 15, characterised in that the frictionally withholding device comprises one or more bars with rounded edges over which the film is dragged while following an adjustable bow-

length of the edge, and said bar or bars and the length of travel in contact with the edge or edges is adapted to prevent P2 wholly solidifying.

17. A method according to claim 15, characterised in that the cooling to the said state is carried out by air-lubricated engagement of the film with a cooling body of controlled temperature.

18. A method according to claim 17, characterised in that the cooling is carried out with the film in air-lubricated engagement with two heating bodies, one on each side of the film, the spacing between said heating bodies preferably being adjustable.

19. A method according to claim 2, characterised in that the said further stretching is carried out in the same longitudinal direction as the hot stretching of the film.

20. A method according to claim 19, characterised in that by a suitable selection of the conditions for the different stretching processes, and optionally by addition of a finely dispersed fracture-promoting material to the extruded blend, the longitudinal orientation after full solidification is adapted to produce locations of rupture of the P1 fibrils and in connection with such rupture extra orientation of P2 in and around the said locations, the locations being generally extended in a linear fashion at an angle to the direction of orientation.

21. A method according to claim 19, characterised in that the said further stretching is carried out at around 50°C or at a lower temperature.

22. A method according to claim 19, characterised in that in succession to said further stretching, transverse stretching is carried out while P1 and P2 are solid, preferably under allowance of a simultaneous longitudinal contraction.

23. A method according to claim 22, characterised in that longitudinal contraction is achieved by forming transverse pleats in the film prior to the transverse stretching, and the latter is carried out by means of a tenter frame.

24. A method according to claim 2, characterised in that the said further stretching is carried out transversely of the preceding longitudinal orientation of the film, preferably while the film is allowed to shrink in said longitudinal direction.

5 25. A method according to claim 24, characterised in that the shrinking is achieved by forming transverse pleats in the film prior to the transverse stretching, and the latter is carried out by means of a tenter frame.

10 26. A method according to claim 1, characterised in that a minor surface layer is coextruded on at least one side of the blend to enhance bonding properties and/or modify frictional properties of the film.

27. A method according to claim 1, characterised in that P1 consists of polypropylene polyamide or polyethylene terephthalate, and P2 mainly consists of a propylene copolymer or polyethylene.

15 28. A method according to claim 27 in which said polypropylene for P1 is a crystalline copolymer of propylene.

29. A method according to claim 27 or 28 in which the polyethylene is a crystalline copolymer of ethylene, preferably linear low density polyethylene.

20 30. A method according to claim 1, characterised in that after the end of the mentioned steps, the film, which exhibits a uniaxial or unbalanced orientation, is laminated to one or more similarly or differently manufactured films of uniaxial or unbalanced biaxial orientation, whereby the films are arranged so that their main directions of orientation cross each other.

25 31. A method according to claim 1, characterised in that additionally to the mentioned steps the film is cut into narrow longitudinally oriented tapes.

30 32. A method of forming a film or sheet of thermoplastic polymer alloy in which there is formed an intimate blend of polymer material P1' and polymer material P2', the blend is extruded through a die and the extruded film is stretched after extrusion in which the flow passage through the die

comprises an exit orifice having an exit gap, characterised in that upstream from the exit orifice there is provided a grid chamber comprising one or more grids through which the blend passes, the grid or grids having at least 4 (in the longitudinal sections perpendicular to the main surfaces of the flow) closely spaced lamellae having walls extending several millimetres in the direction of the flow, and, between the lamellae apertures of a size selected to reduce the average size of the dispersed phase of P1' or P2' in the blend, the grid or grids being located at a position in the chamber where the gap is wider than the said exit gap, the grid chamber further comprising a gap reduction portion between the screen and the die exit wherein the gap through which the blend flows is reduced at least part way to the gap of the die exit.

33. A method according to claim 32 characterised in that the lamellae in each such grid are slanted so that each forms an angle between about 10 to about 70° to the major surface of the blend flow entering the grid.

34. A method according to claim 33, characterised in that the major lamellae in each such grid are substantially planar.

35. A method according to claim 33, in which the lamellae are substantially parallel to the flow as it enters the grid.

36. A method according to claim 34, characterised by at least two such grids which mutually are oppositely slanted in relation to the direction of the blend flow entering the grid.

37. A method according to claim 32, characterised in that there is coextruded a surface layer at least on one side of the blend flow, preferably before this flow meets the grid or grids.

38. A method according to claim 32 characterised in that P1' and P2' are incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised either by use of an alloying agent or mechanically by sufficient mixing and attenuation, and P2' in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1', and preferably but not necessarily the mechanically

determined melting point of P1' is at least about 20°C higher than that of P2', and further by adaptations of rheological conditions, percentages of the components, and conditions for mixing and extruding a dispersion of microscopically fine fibrils or fibril network of P1' surrounded by P2' is formed in the alloy, whereby each fibril extends mainly in one direction and generally has a thickness around or lower than 5µm, preferably around or lower than 1µm, and still more preferably around or lower than 0.1µm and width at least 5 times its thickness, and further characterised in that the film is stretched after at least P1' has been solidified.

39. A method according to claim 38, characterised in that the said stretching is transverse to the direction of the fibrils, and preferably the film is allowed to contract in the direction of the fibrils during said stretching.

40. A method according to claim 38, characterised in that possibilities for contraction are introduced by a preceding fine transverse pleating of the film.

41. A method according to claim 40, characterised in that the step of stretching transverse to the direction of the fibrils is preceded by stretching in the direction of the fibrils while the latter are solid.

42. A method according to claim 38, characterised in that P1' consists of polypropylene, polyamide or polyethylene terephthalate, and P2' mainly consists of a propylene copolymer or polyethylene.

43. A method according to claim 42 in which the polypropylene for P1 is a crystalline copolymer of propylene.

44. A method according to claim 42 or 43 in which the polyethylene is a copolymer of ethylene, preferably linear low density polyethylene.

45. A method according to claim 38, characterised in that the film is given a strong uniaxial or unbalanced biaxial orientation, and subsequently the film is laminated to one or more similarly or differently manufactured film of uniaxial or unbalanced biaxial orientation, whereby the films are arranged so that their main directions of orientation cross each other.

46. A method according to claim 38, characterised in that subsequently the film is cut into narrow longitudinally oriented tapes.

47. A method according to claim 32 characterised in that P1' is chosen to exhibit desirable barrier properties, and P1' and P2' are
5 incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised either by use of an alloying agent or mechanically by sufficient mixing and extension, and preferably but not necessarily the mechanically determined melting point of P1' is at least about 20°C higher than that of P2', and further by adaptations of rheological
10 conditions, percentages of the components, and conditions for mixing and attenuation a dispersion of microscopically fine fibrils or fibril network of P1' surrounded by P2' is formed in the alloy whereby each fibril extends in one main direction, has a thickness around or lower than 5µm, preferably around or lower than 1µm, and has a width at least 5 times its thickness.

15 48. A method according to claim 32 characterised in that P1' and P2' are incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised either by use of an alloying agent or mechanically by sufficient mixing and extrusion and preferably but not necessarily the mechanically determined melting point of P1' is at least
20 about 20°C higher than that of P2', and further by adaptations of rheological conditions, percentages of the components, and conditions for mixing and attenuation a dispersion of microscopically fine fibrils or fibril network of P1' surrounded by P2' is formed in the alloy, whereby each fibril extends mainly in one direction, has a thickness around or lower than 5µm, preferably
25 around or lower than 1 µm, and has width at least 5 times its thickness, and further characterised in that there is added a volatile expansion agent prior to or during the extrusion, which agent is soluble in P2' but generally not in P1', whereby expansion takes place after extrusion.

49. An extruded oriented film which is in the form of a
30 crosslaminated, in which it is laminated to another oriented film, whereby the main directions of orientation cross each other, or is in the form of a rope, twine or woven-tape products, the film comprising a layer of alloy of at least

two polymers P1 and P2, which both are at least partly crystalline at temperatures less than 100°C, wherein P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1, and the alloy comprises a dispersion of microscopically fine fibrils or
5 fibril network of P1 surrounded by P2, wherein each fibril extends mainly in one direction and generally has width and thickness wherein the mean of these two dimensions is around or lower than 5µm, characterised in that

- 10 a) the P1 fibrils are flat and generally parallel with the main surfaces of the film with thicknesses generally around or lower than 1 µm and width at least 5 times the thickness, and/or
b) the oriented film exhibits locations of rupture of the P1 fibrils, which locations each extend in a generally linear fashion across the film at an angle to the direction of orientation.

15 50. A film according to claim 49, characterised by a minor coextruded surface layer on at least one side of the alloy layer to enhance bonding properties and/or modify frictional properties of the film.

51. A film according to claim 49, characterised in that P1 consists of polypropylene polyamide or polyethylene terephthalate, and P2 mainly consists of a propylene copolymer, or polyethylene.

20 52. A film according to claim 51 in which the polypropylene is a crystalline copolymer of propylene.

53. A film according to claim 51 in which the polyethylene is a copolymer of ethylene, preferably linear low density polyethylene.

25 54. An extruded film according to claim 49 which is a crosslaminated.

55. A film according to claim 49 in the form of rope, twine or woven-tape products.

30 56. An extruded film comprising a layer of an alloy of at least two polymers P1 and P2, which both are at least partly crystalline at temperatures under 100°C and are incompatible to such an extent that they exist as separate phases in the final film but are compatibilised sufficiently for practical purposes, comprising a dispersion of microscopically fine fibrils

or fibril network of P1 surrounded by P2, wherein each fibril extends mainly in one direction, characterised in that the fibrils of P1 are flat and generally parallel with the main surfaces of the film with thicknesses generally around or lower than 1 μm , and width at least 5 times the thickness and further
5 characterised in that P1 is chosen to exhibit desirable barrier properties and further characterised by comprising, in longitudinal cross-section perpendicular to the main surfaces of the film, at least 4 die lines.

57. A film according to claim 56, characterised by a minor coextruded surface layer on at least one side of the alloy layer to enhance
10 bonding properties and/or modify its frictional properties.

58. A film according to claim 56, characterised in that P1 consists of EVOH, a vinylidene chloride copolymer or polyamide.

59. An extruded film according to claim 56 which is uniaxially or biaxially oriented and is laminated to another oriented film, whereby the main
15 directions of orientation cross each other.

60. A cellular expanded film made by extrusion in the presence of an expansion agent, characterised in that the film is made from an alloy of at least two polymers P1 and P2, which both are at least partly crystalline at temperatures under 100°C, the alloy comprising a dispersion of
20 microscopically fine fibrils or fibril network of P1 surrounded by P2, whereby each fibril extends mainly in one direction and is flat with thicknesses generally around or lower than 1 μm , and width at least 5 times the thickness.

61. A film according to claim 60 which is uniaxially or biaxially oriented and is laminated to another film, whereby the main directions of
25 orientation cross each other.

62. A film according to claim 60 in the form of rope, twine or woven-tape products.

63. A film according to claim 60 in the form of split fibre products.

64. A film according to any of claims 60 to 63 in which wherein P2
30 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1.

65. A film according to any of claims 56 to 65 in which P2 is a copolymer of propylene or polyethylene, preferably a copolymer of ethylene and another alphaolefin, preferably LLDPE.

5 66. A film according to any of claims 56 to 66, in which, in the alloy, the weight proportion of P1 is in the range 5 to 75 %.

67. Apparatus for extruding thermoplastic material comprising a die having an exit orifice through which the molten material flows and stretching means for stretching the material after it is extruded by at least two steps, in the first of which the material is stretched longitudinally by first stretching
10 means whilst at a high temperature, and in the second of which the material is stretched longitudinally by second stretching means at a lower temperature, comprising also means for cooling the extruded material between the two stretching means, said cooling means comprising a frictional device arranged for contact with the extruded material,
15 characterised by further comprising stretching means downstream from said second stretching means, and additional cooling means between said second stretching means and said further stretching means.

68. Apparatus according to claim 67 in which the said frictional device is provided with holes or is made of microporous metal for inwards or
20 outwards passage of air whereby over or under pressure of air is provided to control the friction between the device and the material.

69. Apparatus according to claim 67 or 68 comprising a shock cooling part upstream of the frictional device past which the extruded flow passes and which is cooled by a flow of cooling medium through its interior.

25 70. Apparatus according to claim 69 which further comprises heating means between the shock cooling means and the frictional device, for controlled heating of the material.

71. Apparatus according to claim 70 in which the heating means comprises a pair of fixed heating blocks arranged on opposite sides of the
30 extruded material.

72. Apparatus according to any of claims 67 to 71 in which the die has a grid chamber upstream from the exit orifice comprising one or more grids through which the extrudate passes, the grid or grids being located at a position in the chamber where the gap is wider than said exit orifice gap, the
5 grid chamber further comprising a gap reduction portion between the grid or grids and the exit orifice wherein the gap is reduced at least part way to the gap of the exit orifice.

73. Apparatus for extruding thermoplastic material comprising a die having an exit orifice through which the molten material flows and stretching
10 means for stretching the material after it is extruded by at least two steps, in the first of which the material is stretched longitudinally by first stretching means whilst at a high temperature, and in the second of which the material is stretched longitudinally by second stretching means at a lower
15 temperature, comprising also means for cooling the extruded material between the two stretching means, said cooling means comprising a frictional device arranged for contact with the extruded material, characterised in that there is provided a grid chamber upstream from the exit orifice comprising one or more grids through which the extrudate passes, the
20 grid or grids being located at a position in the chamber where the gap is wider than said exit orifice gap, the grid chamber further comprising a gap reduction portion between the grid or grids and the die exit wherein the gap is reduced at least part way to the gap of the exit orifice.

74. Apparatus according to claim 72 or 73 in which each such grid has walls extending several mm in the direction of the flow.

25 75. Apparatus according to any of claims 73 to 74 in which the major walls in each such grid are substantially planar and are slanted so that each forms an angle between about 10 to 70° to the major surface of the extrudate flow entering the grid.

30 76. Apparatus according to claim 75 in which said angle and the wall thickness and distances between the walls are such that, in a

longitudinal section of the die perpendicular to the main surfaces of the extrudate flow as this enters the grid, there are at least four such walls.

77. Apparatus according to claim 75 or 76 which comprises at least two such grids which are slanted in opposite directions to one another.

5 78. Apparatus according to any of claims 67 to 77 comprising means for coextruding a surface layer at least on one side of the extrudate.

79. Apparatus according to any of claims 67 to 78 comprising means for transverse stretching of the extruded film downstream of the second stretching means.

10 80. Apparatus according to claim 79 in which upstream of the transverse stretching means there is a longitudinal pleating device, preferably comprising a pair of rubber belts between which the extruded material passes.

81. Apparatus according to any of claims 79 to 80, in which the transverse stretching means comprises a tenterframe including an oven.

82. Apparatus according to claim 81, in which said oven comprises fixed heated blocks arranged on opposite sides of the material, provided with heating means, preferably electrical heating means.

20 83. Apparatus according to claim 82, which further comprises a cooling block on at least one side of the material, downstream of the heating blocks, said cooling block being provided with a channel for passage of cooling air, preferably in which said channel is in fluid communication with the surface of the block facing the extruded material, by virtue of forming the cooling block from microporous metal.

25 84. Apparatus according to claim 82 or 83, in which said heating blocks are formed of microporous metal in fluid contact with channels for passage of heated air, whereby heated air exits the blocks from the surfaces facing the material passing therebetween, to lubricate passage of the material therebetween.

30 85. Apparatus according to any of claims 67 to 72, in which the further stretching means is a longitudinal stretching means and the

apparatus preferably comprises pleating means for imposing transverse pleats in the material prior to said longitudinal stretching.

86. Apparatus according to claim 85, including a laminating station, in which a second sheet material is laminated to the extrudate, said
5 laminating station preferably being upstream from the longitudinal cold stretching means.

87. Apparatus according to claim 86, in which the extrusion die is a circular die for extruding a tube of material, and which further comprises helical cutting means downstream of the said second stretching station, and
10 upstream of the laminating station, in which the tube of material is helically cut and two plies of the extruded material are laminated to one another with their main directions of orientation arranged at an angle to one another.

88. Apparatus according to any of claims 67 to 86 in which the extrusion die is a flat die.

89. Apparatus for extruding thermoplastic material comprising a die
15 having an exit orifice through which the molten material flows and stretching means for stretching the material after it is extruded, characterised in that the die has a grid chamber upstream from the exit orifice comprising one or more grids through which the extrudate passes, the grid or grids being
20 located at a position in the chamber where the gap is wider than said exit orifice gap, the grid chamber further comprising a gap reduction portion between the grid or grids and the exit orifice wherein the gap is reduced at least part way to the gap of the exit orifice and the or each grid comprises at
25 least 4 (in the longitudinal sections perpendicular to the main surfaces of the flow) closely spaced lamellae having walls extending several mm in the direction of flow of molten material and, between the lamellae having apertures through which the molten material can flow.

90. Apparatus according to claim 89 in which the lamellae in each such grid are slanted so that each forms an angle between about 10 to 70°
30 to the major surface of the extrudate flow entering the grid.

91. Apparatus according to claim 90 in which the lamellae are substantially planar and preferably are substantially parallel to the flow as it enter the grid.

5 92. Apparatus according to claim 90 or 91 which comprises at least two such grids which are slanted in opposite directions to one another.

93. Apparatus according to claim of claims 89 to 92 comprising means for coextruding a surface layer at least on one side of the extrudate.

94. Apparatus according to any of claims 89 to 93 in which the extrusion die is a circular die for extruding a tube of material.

10 95. Apparatus according to any of claims 89 to 93 in which the extrusion die is a flat die.

96. A film according to claim 49a, 56 or 60 in which the widths of the fibrils are at least 10 times the thickness.

15 97. A film according to claim 49 which , in longitudinal cross-section perpendicular to the main surface of the film, comprises at least 4 die lines.

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**METHOD OF MANUFACTURING AN ORIENTED FILM FROM ALLOYED
THERMOPLASTIC POLYMERS, APPARATUS FOR SUCH
MANUFACTURE AND RESULTING PRODUCTS**

The invention concerns novel, oriented morphologies and special kind
5 of orientation in a polymer alloy and is conceived with special view to film
used for crosslamination and film used for manufacture of rope, twine or
woven tape fabrics.

One objective is to improve the mechanical performance of the
product at elevated temperatures and in this connection improve the
10 heatseal properties.

Another objective is formation of a morphology which optimizes creep
properties without sacrificing energy absorption properties.

A third objective is the formation of a morphology comprising
microscopic or submicroscopic thin ribbon-formed fibrils, which promote
15 barrier properties or enable an improved action of expansion agents.

A fourth objective is, based on suitable morphology, to facilitate and
improve transverse orientation of thermoplastic film, especially with a view to
a rationalised, in-line manufacture of crosslaminates.

The manufacture of rope, twine, and woven technical products, all
20 based on uniaxially oriented, slit film, can in many instances be rationalised
if the film gets suitable heatsealing properties. Furthermore, for many uses
there exists a need to achieve improved tensile energy absorption in such
products without being forced to use high-cost polymers, such as
polyamides. Finally, the formation of a fine cellular expanded structure in
25 oriented tape may open new market opportunities.

Crosslaminates from oriented thermoplastic polymer films present the
advantage that suitable combinations of different types of high strength
properties can be achieved at relatively low cost, e.g. based on cheap
polyolefins such as polyethylene or polypropylene. The cost saving comes
30 from reduction of the gauge, and thereby of the raw material consumption,
but this must be weighed against the cost of the orientation, stretching and

oriented on the bias, and continuously laminating two such webs between rollers such that the directions of orientation in the two webs are oppositely biased. The lamination can be an extrusion lamination or, if the films have been coextruded, may be by heat with the use of lamination layers.

5 The patent basis for this manufacturing method and resultant product is GB-A-0,192,976 and GB-A-0,816,607 (and corresponding patents).

 In another process sequence used industrially, there are first coextruded tubular films each with a main layer consisting of a blend of selected polymers and minor layers partly serving as lamination layers and
10 partly as heat-seal layers for the final laminate. During the haul-off from the extrusion device the tubular films are given a strong melt orientation mainly in the direction of extrusion. Without any further stretching carried out, the tubes are helically cut, continuously crosslaminated by heat and pressure in a roller apparatus and, in the same apparatus, biaxially oriented. The
15 transverse component of this orientation is carried out between several pairs of intermeshing grooved rollers. The patent basis for this manufacturing method is GB-A-1,526,722 (and corresponding patents). Since this is particularly relevant in connection with the present invention, the main method claim shall hereinafter be paraphrased.

20 It concerns a method of making a laminate comprising extruding at least two layers of molten polymeric blend, each layer comprising a blend of polymers that are incompatible to such an extent that on solidification each comprises a dispersion of particles of one polymer in a matrix of another polymer, attenuating each layer while molten before, during or after
25 extrusion to distort the particles in each layer into a fibrillar grain structure having a predominant direction of splittability after solidification into a film, bonding the layers with the said predominant directions transverse to each other, solidifying the layers if they are not already solidified, and biaxially orienting the resulting laminate by orienting it in substantially uniaxial steps.
30 Hereby the biaxial orientation is conducted at a temperature sufficiently low to maintain the predominant direction of splittability of each film and the

However, the two first mentioned inventions suffer from the drawback that special equipment is needed in the company which carries out the conversion of the film to heat-sealed products (e.g. bags) and thus can be a serious marketing limitation for the manufacturer of the film. In the last
5 mentioned invention the solution concerns features only in the composition and manufacturing process of the cross laminated film, but further improvements of the heat-sealing properties of such film are still highly desirable. As mentioned above this is also an objective of the present invention.

10 Another serious problem has been the flimsiness of a crosslaminated which is made in the low gauges which are allowed by the improvements of strength properties. Stiffness and resilience is particularly important in the manufacture and/or automatic handling of bags. This problem is partly solved in the above mentioned US-A-5,626,944 which discloses a ribbed
15 structure of the laminate in which each rib has a cross-section like a very shallow U, formed by a special execution of the transverse stretching between grooved rollers.

A more radical solution is disclosed in WO-A-02/102592 in which at least one of the films of the laminate forms flutes like the flutes in corrugated
20 paperboard, generally with a wavelength between about 1 mm and 3 mm.

The present invention can be carried out either additionally to or supplementally to the process steps which are referred to in the foregoing description of known crosslamination technology.

US 2,262,989 describes a method of forming a sheet of thermoplastic
25 material, in which the material is extruded through a die and is stretched after extrusion, in which the flow passage through the die comprises an exit orifice having an exit gap. In the exit orifice there is a mixing screen, comprising a plate having a plurality of circular apertures varying.

In JP-A-57-110422 (1982) there is described a method of forming a
30 plate of thermoplastic polymer alloy, in which the mixing chamber comprises a static mixer formed from several devices comprising small plate mounted

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on supports, and oriented at an angle to the direction of flow. The objective of the static mixer is to avoid die lines in the resultant plate.

In EP-A-0337719, there is described a method of forming a thermoplastic film composed of a matrix of a first thermoplastic resin and substantially continuous tapes formed of a second thermoplastic resin
5 having barrier properties dispersed in the matrix.

A first aspect of the present invention is based on the concept that the fibrillar grain structure mentioned above is given a strong orientation at a temperature at which the fibrils are solid while the surrounding polymer
10 material (which in solid state also normally is at least partly crystalline) is molten. In particular the method involves orientation over a frictional surface while the part-molten film is hauled off from the extrusion device, although it also is possible first to solidify, later partly re-melt the film and then perform the stretching. After this hot stretching and after solidification of the entire

film composition with mainly the fibrils oriented, the film is preferably further stretched at a lower, preferably much lower, temperature. Such subsequent cold stretching will normally rupture the fibrils or fibril network at microscopic intervals along the length of the fibrils, while the film material on the whole remains intact. This state has been found to provided improved yield tension and creep properties without adverse effects on tensile energy absorption and behaviour of the film under shock, e.g. shock-tearing or shock-puncturing, a discovery which is further described below.

The feature that the higher melting fibrils have a much higher degree of orientation than the surrounding lower melting polymer material also provides the film with improved strength properties at elevated temperatures, and in particular improved heat-sealing properties. It has been found that an essential part of the strength-generating orientation in P1 is maintained above the melting point of P2, see in this connection the shrink-testing of examples of the invention below.

The first aspect of the present invention is a further development of the method and apparatus, which the inventor has disclosed in WO03/033241. This earlier case discloses extrusion of a tubular film over an annular frictional device of controllable temperature, adapted to produce longitudinal orientation while the temperature in the flow is kept within or slightly above the crystallisation range of the flow of polymers. An embodiment of that invention is characterised in that the polymer flow contains a blend of at least two compatible or compatibilised polymers, and the main proportion of the orientation takes place while one is predominantly in crystalline state and the other is predominantly in molten state. The friction which controls the orientation, can be adjusted by air lubrication with air pressed through holes in the frictional device or through microporous metal, or alternatively by sucking the flow against the frictional device. The surface which the flow contacts may also have a grooved pattern, the

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agent or mechanically by sufficient mixing. P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1. By adaptations of rheological conditions, percentages of the components, and mixing and stretching conditions the alloy is produced which is a
5 dispersion of microscopically fine fibrils or fibril network of P1 surrounded by P2, wherein each fibril extends mainly in one direction and generally has width and thickness such that the mean of these two dimensions is around or lower than 5µm. The stretching takes place partly by draw-down after extrusion of the film while both components are at least partially molten, and
10 partly by a later step to form strong orientation.

The features which characterise the first aspect of the invention are as follows: after said draw-down (also termed attenuation or hot attenuation, below) the film is hot stretched while P1 is in solid state and P2 molten or semimolten to selectively orient P1, while limiting the orientation to an extent
15 which allows the film to become at least 25% elongated in the direction of orientation by slow drawing at 20°C whereby the hot stretching is carried out by drawing the film over a frictionally withholding device (frictional device).

"Selectively orient" does not mean that P2 does not become oriented, it always will do so to some extent.

20 After the described hot stretching the film is preferably further stretched while both components are solid (cold stretching). The resultant orientation is hereby preferably limited so that the product film can be elongated at least 25% in any direction by slow drawing at 20°C. "Slow drawing" refers to stretching of narrow, e.g. 15mm wide specimens at a
25 velocity corresponding to about 50% elongation per minute. This characteristic is also referred to as the elongation at break.

In order for polymer P2 to exert its energy absorbing properties properly the coefficient of elasticity E for this polymer material in its unoriented state must as mentioned be at least 15% lower than E for
30 polymer material P1 in the unoriented state of P1. However, it will in many cases be more preferable to choose P2 with a still lower E seen in relation to

P1, e.g. such that its E is 25% lower than that of P2, 50% lower or even lower than this.

For the sake of good order it should be mentioned that part of the blending may take place already during manufacture of the polymerisates
5 before the polymers P1 and P2 have been isolated, or thereafter in a separate manufacturing location or step.

Preferably polymers P1 and P2 should be compatible or compatibilised to such an extent that the final film will not shown any tendency to develop internal voiding upon repeated flexing. As mentioned
10 above this compatibilisation can be by the use of an alloying agent, or more economically by making the cross-sectional dimensions of the fibrils sufficiently small, since the tendency to internal voiding is dependent on these dimensions.

Suitable examples of polymers for use in the invention are:

15 P1: **propylene polymers including crystalline copolymers of propylene, or polyamide, or polyethylene terephthalate, and**

P2: **propylene copolymer, or ethylene polymers including crystalline copolymers of ethylene, e.g. with other alphaolefins.**

When the film is made for manufacture of a crosslaminate, it is
20 preferably a coextruded film which, on the side which will face another ply in the laminate, has a lamination layer selected to facilitate or control the bonding. Furthermore it is most advantageous that the outermost plies of the laminate each will comprise a coextruded modifying layer forming a surface of the entire laminate, selected to facilitate and/or improve
25 heatsealing of the laminate and/or modify its frictional properties or printing characteristics.

It would appear that an oriented film as described above could be made by a simple modification of the process which is described in US-A-3,949,042 (Utz). According to this patent a crystalline polymer is solidified
30 by cooling while it passes through the exit part of an extrusion device, and is oriented by the pull of the haul-off means as it leaves the exit orifice. Further

with the highly attenuated, very flat fibrils, can provide crosslaminates and slit-film products with improved strength properties, independent of the special features of the first aspect. Furthermore it can be used to manufacture crosslaminates, slit film and other film products, e.g. films with a fine cellular, expanded structure. Finally the extended, very flat polypropylene microfibrils, alloyed into a film of linear low density polyethylene (LLDPE) can render a film highly oil resistant, while very flat polyamide microfibrils, also alloyed with LLDPE can do the same and further provide good barrier properties against oxygen. This second aspect of the present invention shall be dealt with in more detail below.

Going back to the first aspect of the invention, this can be carried out in different sequences of steps. In one sequence the film is after the extrusion and the hot-draw-down, while both P1 and P2 are molten, cooled to solidify both, and in a later step heated in air-lubricated engagement with a heating body of controlled temperature to melt at least in part P2, while keeping P1 solid. Immediately thereafter while P2 is still at least partially molten and P1 is solid, the film is subjected to the selective orientation of P1, i.e. hot stretching and then solidification of P2. The frictional device preferably comprises one or more bars with rounded edges over which the film is dragged while following an adjustable arc of the edge, and the bar or bars are maintained at a temperature which prevents the film from sticking to the edge or edges. The length of travel in contact with the edge or edges is adapted to prevent P2 wholly solidifying. An example of such frictional device, suited for film extruded in flat form, is shown in fig. 5 herein, and another example, suited for tubular film, is shown in fig. 6 of the mentioned WO03/033241.

At least the process steps from and including extrusion to and including the solidification of P2 are preferably carried out in-line, whereby the line also comprises a hold-back device, e.g. similar to one of those mentioned above, acting between the cooling and the subsequent heating,

temperature-fine-adjustment part (117). Each one can be constructed like the said frictional device (118) or as shown in fig. 6, both from the WO document.

5 The heating is preferably carried out with the film in air-lubricated engagement with two heating bodies, one on each side of the film. The spacing between said heating bodies should preferably be adjustable. This is also illustrated in the drawing fig. 4 herein.

10 In an alternative sequence of steps, cooling of the film immediately after the extrusion is limited to solidification of P1, while P2 is kept at least partially molten. Immediately thereafter, the selective orientation of P1 over a frictionally withholding device is carried out with P1 in a crystalline state and P2 at least partially molten. This sequence of steps is shown in the flowsheet fig. 2. Also in this case, the frictionally withholding device may
15 comprise one or more bars with rounded edges over which the film is dragged while following an adjustable arc of the edge, whereby the temperature of the bar or bars and the length of travel in contact with the edge or edges is adapted to prevent P2 wholly solidifying.

The cooling to the state in which P1 has become solid while P2 remains at least partially molten, is preferably carried out by air-lubricated
20 engagement of the film with a cooling body of controlled temperature. Also in this case the film should preferably be in air-lubricated engagement with two temperature controlling bodies one on each side of the film. The spacing between said heating bodies should preferably be adjustable.

25 As regards the orientation after full solidification of both components, the cold stretching, at least in a first step is preferably carried out in the same longitudinal direction as the previous hot stretching of the film. By a suitable selection of the conditions for the different stretching processes, and optionally by addition of finely dispersed fracture-promoting material to the extruded blend, the longitudinal cold stretching is preferably adapted to
30 produce locations of rupture of the P1 fibrils and, in connection with such rupture, extra orientation of P2 in and around the said locations. Hereby the

tubular, spiral cutting may be used as it normally is done (see the introduction).

5 The second aspect of the method according to the present invention shall now be described more exactly. It concerns a method of extruding and attenuating thermoplastic polymer material, which comprises an intimate blend of polymer material P1' and polymer material P2', through a die and stretching the extruded film to form a film or sheet of an alloy, in which the flow passage through the die comprises an exit orifice having an exit gap, and the method is characterised in that upstream from the exit orifice there is
10 provided a grid chamber comprising one or more grids through which the blend passes. The grids have at least 4 (in the longitudinal sections perpendicular to the main surfaces of the flow) closely spaced lamellae having walls extruding several mm in the direction of the flow. The grid or grids have apertures of a size selected to reduce the average size of the
15 dispersed phase of P1' or P2' in the blend. The grid or grids are located at a position in the chamber where the gap is wider than the said exit gap, and the grid chamber further comprises a gap reduction portion between the screen and the die exit wherein the gap through which the blend flows is reduced at least part way to the gap of the die exit.

20 The preferred features of this die has already been clearly described in connection with the first aspect of the invention.

For most uses of the film product there is preferably coextruded a surface layer at least on one side of the blend flow. This is best done before the flow meets the grid or grids, since the latter then can help to
25 compatibilize the blend of the main layer and the surface layer.

Independently of the first aspect of the present invention, this second aspect can be used to improve strength properties of alloyed, oriented film.

An embodiment of this method is characterised by the following features: P1' and P2' are incompatible to such an extent that they exist as
30 separate phases in the final film, but are compatibilised sufficiently for practical purposes either by use of an alloying agent or mechanically by sufficient mixing and attenuation, and P2' in its unoriented state at 20°C exhibit a coefficient of elasticity (E) which is at least 15% lower than E of P1'. Preferably but not

and they may be converted to split fibre network e.g. by rubbing action as known in the art of making split fibres (fibrillated film).

As it already appears from the foregoing, the new methods described above can be used to manufacture the following novel products A, B and C.

- 5 All three are especially conceived for use in crosslaminates, A and C also especially for rope, twine and woven-tape products, and product C is further especially conceived for conversion to split fibre - (fibre-from-film) products.

In the following description reference is made to polymers P1 and P2, i.e. as described above in connection to the first aspect of the invention.

- 10 The description however relates equally to blends of P1' and P2' and products produced therefrom by the second aspect.

Product A: Known features are as follows:

- It is an extruded film made from an alloy of at least two polymers P1 and P2, which both are at least partly crystalline under about 100°C, and are
15 incompatible to such an extent that they exist as separate phases in the film, but are compatibilised sufficiently for practical purposes either by use of an alloying agent or mechanically by sufficiently mixing. P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1, and the alloy is formed substantially as a dispersion of
20 microscopically fine fibrils or fibril network of P1 surrounded by P2. In this morphology, each fibril extends mainly in one direction and generally has width and thickness which as an average between these two dimensions is around or lower than 5µm.

- Characterising features are the following alternatives, which also can
25 be combined:

- a) the P1 fibrils are flat and generally parallel with the main surfaces of the film with thicknesses generally around or lower than 1 µm and a width at least 5 times the thickness,
b) the oriented film exhibits locations of rupture of the P1 fibrils,
30 which locations each have a generally linear extension across the film angularly to the direction of orientation.

As important examples P1 may consist of polypropylene including crystalline copolymers of propylene, or of polyamide, or polyethylene terephthalate, and P2 may mainly consist of a propylene copolymer, or polyethylene including crystalline copolymers of ethylene, preferably linear
5 low density polyethylene.

There should normally be a minor coextruded surface layer on at least one side of the alloyed layer to enhance bonding properties and/or modify the frictional properties.

Product B, an expanded product, exhibits the following known
10 features:

It is an extruded film made from an alloy of at least two polymers P1 and P2, which both are at least partly crystalline under about 100°C, and are incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised sufficiently for practical purposes either by use of
15 an alloying agent or mechanically by sufficient mixing and attenuation. The alloy is formed substantially as a dispersion of microscopically fine fibrils or fibril network of P1 surrounded by P2, whereby each fibril extends mainly in one direction.

Characterising features are as follows:
20 the fibrils of P1 are flat and generally parallel with the main surfaces of the film with thicknesses generally around or lower than 1 µm and a width at least 5 times the thickness, and further more P1 is chosen to exhibit desirable barrier properties.

The barrier forming polymer P1 may e.g. consist of EVOH, or
25 polyvinylidene chloride including copolymers of vinylidene chloride, or polyamide.

There should normally be a minor coextruded surface layer on at least one side of the alloyed layer to enhance bonding properties and/or modify its frictional properties.

30 Product C is a cellular expanded film may be extrusion under use of an expansion agent. It comprises the following novel features:

lubricated engagement with both. The section of each oven part, which is adjacent to the film, consists of microporous metal, the rest of solid metal with one channel system for distribution of the air, and one for heating with circulating hot oil. The channel systems are not shown here, but similar
5 channel systems are shown in figs. 1-5 in WO03/033241. The temperature of the surface of each oven part which is adjacent to the film (211) is exactly controlled. The heating by circulating oil may be substituted by electrical heating. The air may be preheated, but normally its temperature will become precisely adjusted by the passage through the microporous metal, without
10 any preheating.

The double arrows (215) indicate that the spacing between the two oven parts are mutually adjustable to optimize the heating and the effect of the air lubrication. Also in this case, the adjustment preferably takes place through springs which are under an adjustable tension. There are two
15 reasons for using two oven parts (213) and (214) instead of only one (213). One is to achieve the most efficient heating, the other to prevent that the film forms longitudinal pleats as a result of some longitudinal stretching, which will occur at the downstream end of part (213) where the film is most warm. Without the other oven part (214) this stretching would be accompanied by a
20 transverse contraction, which would cause some pleating at the upstream end of part (213). Another but less efficient way to counteract pleating, is to make the microporous surface or surfaces which engage with the film slightly rounded, e.g. with radius of curvature 1-2 meters, and in fig 4. this is also indicated.

25 The special oven consisting of parts (213) and (214) corresponds to the temperature-fine-adjustment-part (117) in figs. 3-5 of WO03/033241, relating to the hot drawing of tubular film. However, to carry out the process shown in the flowsheet, present fig. 1, these figs. 3-5 of WO03/033241 must be supplemented by a suitable "hold-back" device between the shock-
30 cooling part (116) and the temperature-fine-adjustment-part (117). This hold-back device must exert a controlled frictional resistance in order to

second pull roller (223). Rollers (222 and 223) are watercooled steel rollers which solidify P2 (step 6).

Step 7, the cold stretching, i.e. orienting while both P1 and P2 are solid, may be longitudinal or transverse or a combination of both. Fig. 4 shows longitudinal coldstretching. Preferably the film is maintained at a somewhat elevated temperature, e.g. about 50°C, when it leaves roller 224. It is pulled by the cooled, driven rollers (225 and 226). Rubber-coated nip roller (227) forms a nip with the second driven roller (226). The orientation takes place as the film passes and leaves the stretching bar (228). This has a semi-sharp edge, e.g. rounded with radius of curvature of about 0.5mm, over which the film is drawn. By turning of the bar around the centre axis, which is indicated in the drawing, the friction can be varied.

The flowsheet fig. 2 and corresponding drawing of machine line fig. 6 show an alternative route for carrying out the first aspect of the invention.

This does not first solidify both components and then re-melt P2, but goes directly from step 2, melt attenuation, over a step 2a, controlled withholding, to the step (3a) where P1 is solidified while P2 remains molten.

When working with flat film, as shown in fig. 6, the controlled withholding (step 2a) which at the same time performs the controlled melt-attenuation, can conveniently take place by means of the cooled roller (206). This is cooled in order to prevent the film from adhering, but the cooling must not be so strong that a substantial portion of the film solidifies. Therefore the film follows the roller over a very short distance, e.g. 1-2 cm. It is pressed against a roller (206) by means of an air lubricated bar (210) which is generally similar to the bar (210) in fig. 4, but it is essential to make the construction so that the surface which faces the film is no wider than about 5-10 mm. This device may be heated in order to avoid excessive cooling of the film during this step.

Although step 3a in fig. 2 concerns solidification of P1 while keeping P2 at least partially molten, and step 3a in fig. 1 concerns melting of P2 while keeping P1 solid, both steps are preferably carried out with essentially

hereby forms transversely extending thinner lines. The distance between such lines may e.g. be about 0.5-3 mm. Even a small degree of incremental longitudinal stretching will help to produce an even pleating, and furthermore it has been found that the longitudinal stretching zones can act as initiators for the transverse stretching.

As the pleated film leaves the stuffing device, its edges are gripped by holding clamps in endless tenter-chains (see the schematic sketch fig. 7a) which can be a conventional tenterframe construction.

The oven construction is new. In known tenterframes the oven which heats the film by means of hot air extends over practically the entire film surface during the entire tentering process. The novel concept of an oven or ovens for a tenterframe is schematically shown in fig. 7a, and its cross-section is shown in detail in fig. 7b. The oven consists of two parts (235/236 and 237) made from microporous metal each with support parts (239 and 240) made from compact metal. The component (235) and the part (237) are heated to an appropriate, controlled stretching temperature, e.g. 50°C, by means of electrical heating elements (241). Pressurised air is distributed through the channels (242) between parts 235/236 and 237 on the one hand and the support parts (239 and 240) on the other. The component (236) is not heated, and the air lubrication through this will provide cooling while the air lubrication through the adjacent parts (235) and (237) will provide heating.

The heated component (235) is insulated from the cooling component (236) by means of a heat insulating wall (246) of compact material. The cooling component (236) is on the side of each oven which faces its nearest tenterchain.

The spacing between components (235) and (237) is adjustable as indicated by the double arrows (243). This adjustment may be a simple setting of the distance, or may be through springs with adjustable tension. A preferable spacing between (235) and (237) is between about 0.5 and 2mm. Fig. 7c shows the film entering the oven in pleated form (244) at the

right hand side and leaving the oven centered and with the pleats stretched out (245) in the left hand side.

In a conventional tenterframe oven, the film is stretched simultaneously over its full width, and carrying this out evenly requires a rather high temperature, which is generally not adequate in the manufacture of crosslaminates. With the present constructions, when the parameters are properly adjusted to each other, the tentering takes place within a very narrow zone, a necking zone, usually only a few mm wide or less, located on each of the cooling components (236) of the two narrow ovens shown. Due to the air lubrications, the film moves through the ovens in a friction free manner, while the necking zone gradually develops from a position close to each of the chains with holding clamps towards the middle of the film. Thus all film between a narrow oven and the nearest chain (except a narrow zone near the chain) is fully transversely stretched up to the stretch ratio set, while the film between the two narrow, long ovens are not at all transversely stretched. At the downstream end of the two narrow, long ovens, all the film is generally evenly stretched.

When the stretching conditions are properly set, the necking zone will as mentioned be located on the cooling part. This has the effect that the degree of stretching better can be limited, as is generally desired in the manufacturing of crosslaminates, for the purpose of allowing further stretching under impact actions (shock) on the final product.

As it will be understood, the described tenterframe process is not limited to use in the invention as this is defined in the claims, but it is the best way known to the inventor of utilising the invention in the manufacture of crosslaminates. As a further aspect of the invention there is provided a method of transverse stretching of films while pleating using the oven arrangement described above and apparatus therefore.

It is further noted that the morphology achieved by the invention, namely the oriented P1 fibrils in a generally unoriented or only coldstretched P2 matrix, has been found to act stabilising on the tenterframe process.

Shrink testing

The samples which were strain/stress tested (apart from the "as extruded" samples) have also been tested for free shrinkage under a gradual heating from slightly elevated temperatures to 180°C, whereby the readings began at 90°C.

Tested were also three samples (20% PP, 33% PP and 50% PP) which had not been hotdrawn but were cold drawn in ratio 3.00:1 at 20°C. The test results are recorded in figures 12a-d.

The testing was carried out on a heated metal plate of the type used for determination of melting points. The temperature was measured on the plate surface with a thermocouple. The specimens were held towards the plate by an object glass (made for microscopy). Plate, specimens and object glass were carefully lubricated with talc. Two specimens from the same sample were always tested simultaneously, and the values of shrinkage shown in the graphs are averages between the readings from the two specimens.

All of the graphs in figs 12b-d show the shrinkage of two different samples, one hotdrawn at about 130°C, i.e. while the LLDPE clearly was molten, and one hotdrawn at 115°C, i.e. while LLDPE was semisolid.

"Coldrawn 1:1" means not cold stretched at all.

All graphs show three different regions, one below the melting point of LLDPE, a second between this melting point and the melting point of the PP, and a third above the melting point of the PP.

Studying the shrinkage of the 20% PP sample in fig. 12a, there is simply zero shrinkage in the second region, that means the forces of shrinkage exerted by the LLDPE molecules from 90° to 120° essentially eliminated the orientation of the PP molecules. For the corresponding samples with 33% of PP and with 50% PP, the result was not quite but almost the same. Thus, all molecular orientation which essentially contributed to the strength is lost above the melting point of LLDPE. There is some melt orientation left in the PP which causes essential shrinkage above the melting point of PP, but